MEMORANDUM

SUBJECT: **Estimation of Policy-Relevant Background Concentrations of Particulate Matter**

FROM: John Langstaff

TO: PM NAAQS Review Docket (OAR-2001-0017)

DATE: January 27, 2005

The attached technical memorandum describes an analysis of 24-hour average policy-relevant background concentrations of particulate matter.

ESTIMATION OF POLICY-RELEVANT BACKGROUND CONCENTRATIONS OF PARTICULATE MATTER

Technical Memorandum

January 27, 2005

John E. Langstaff

U.S. Environmental Protection Agency
Office of Air Quality Planning and Standards
Air Quality Strategies & Standards Division
Health and Ecosystems Effects Group
Research Triangle Park, North Carolina

Table of Contents

List of Tables

List of Figures

Introduction

For the purposes of the PM Staff Paper (EPA, 2005), policy-relevant background (referred to as "background" in the rest of this document) PM is defined as the distribution of PM concentrations that would be observed in the U.S. in the absence of anthropogenic (man-made) emissions of primary PM and precursor emissions (e.g., VOC, NO_x, SO_2, and NH_3) in the U.S., Canada and Mexico. The reason for defining background in this manner is that for purposes of determining the adequacy of current standards and the need, if any, to revise the standards, EPA is focused on the effects and risks associated with pollutant levels that can be controlled by U.S. regulations or through international agreements with border countries. Thus, as defined here, background includes PM from natural sources and transport of PM from both natural and man-made sources outside of the U.S. and its neighboring countries. Estimating background PM concentrations is important for the health risk analyses and the assessment of ecosystem and visibility effects in the PM Staff Paper.

It is instructive to consider the structure of PM background concentrations from a number of different viewpoints: (1) natural and anthropogenic, (2) local and transported, and (3) baseline and episodic, and we draw upon these concepts in this memorandum. Each of these three views partitions background concentrations into one of two conceptual components. These distinctions are useful in formulating how background concentrations are estimated and elucidating policy relevant issues.

Natural and Anthropogenic Sources

Background levels of PM vary by geographic location and season, and have a natural component and an anthropogenic component. The natural background can be locally generated or be transported large distances, and arises from processes such as: physical processes of the atmosphere that entrain particles (e.g., windblown crustal material, sea salt spray); volcanic eruptions (e.g., sulfates, ash); natural combustion such as wildfires (e.g., elemental and organic carbon, and inorganic and organic PM precursors); and biogenic sources such as vegetation, microorganisms, and wildlife (e.g., organic PM, inorganic and organic PM precursors). The anthropogenic component of background is due to the transport of PM and PM precursors from outside the U.S., Canada and Mexico.

Regional and Transcontinental Transport

PM can be transported long distances from natural or quasi-natural events, as well as anthropogenic emissions, occurring outside the continental U.S., and the PM CD (EPA, 2004) discusses the increasing recognition and understanding of the long-range transport of PM from outside the U.S. The occurrence and location of these long-range transport events are highly variable and their impacts on the U.S. are equally variable. The contributions to background from sources outside of the U.S., Canada and Mexico can be significant on an annual, as well as episodic, basis. Several studies have focused on identifying the origin, sources, and impacts of international transport events from North American and extra-continental sources.

Baseline and Episodic Background

Background concentrations of $PM_{2.5}$, $PM_{10-2.5}$, and PM_{10} may be conceptually viewed in another way as comprised of baseline and episodic components. The baseline component is the contribution from natural sources within the U.S., Canada and Mexico and from transport of natural and anthropogenic sources outside of the U.S., Canada and Mexico that is reasonably well characterized by a consistent pattern of daily values each year, although they may vary by region and season.

In addition to this baseline contribution to background concentrations, a second component consists of more rare episodic high-concentration events over shorter periods of time (e.g., days or weeks) both within North America (e.g., volcanic eruptions, large forest fires) and from outside of North America (e.g., transport related to dust storms from deserts in North Africa and China). Specific natural events such as wildfires, volcanic eruptions, and dust storms, both of North American and intercontinental origin, can lead to very high levels of PM comparable to, or greater than, those driven by man-made emissions in polluted urban atmospheres. Because such excursions can be essentially uncontrollable, EPA has in place policies that can remove consideration of them, where appropriate, from attainment decisions[1].

Geographic Variation in Background

Section 3.3.3 of the PM CD discusses annual average background PM levels, and states that "[e]stimates of annually averaged PRB concentrations or their range have not changed from the 1996 PM AQCD" (CD, p. 3-105). The ranges for $PM_{2.5}$ and PM_{10} are reproduced in Table 1. The lower bounds of these ranges are based on "natural" background midrange concentrations. The upper bounds are derived from the multi-year annual averages of remote monitoring sites in the IMPROVE network (p. 6-44, 1996 PM CD (EPA, 1996)). The ranges for $PM_{10-2.5}$ are derived from the $PM_{2.5}$ ranges and the PM_{10} ranges by subtraction (min PM_{10} – max $PM_{2.5}$, max PM_{10} – min $PM_{2.5}$).

---

[1] There are two policies which allow PM data to be flagged for special consideration due to natural events: the Exceptional Events Guideline (EPA, 1986) and the PM_{10} Natural Events Policy (Nichols, 1996). Under these policies, EPA will exercise its discretion not to designate areas as nonattainment and/or to discount data in circumstances where an area would attain but for exceedances that result from uncontrollable natural events. Three categories of natural PM_{10} events are specified in the natural events policy: volcanic or seismic activity, wildland fires, and high wind dust events. The exceptional events policy covers natural and other events not expected to recur at a given location and applies to all criteria pollutants. Categories of events covered in the exceptional events guidance include, but are not limited to, high winds, volcanic eruptions, forest fires, and high pollen counts. EPA is drafting further guidance concerning how to handle data affected by natural events related to the PM standards.

Table 1. PM CD Estimated Range of Annual Average PM Regional Background Levels (μg/m^3)

	Western U.S.	Eastern U.S.
PM_{10}	4 – 8	5 – 11
$PM_{2.5}$	1 – 4	2 – 5
$PM_{10-2.5}$	0 – 7	0 – 9

There is a distinct geographic difference in background levels, with lower levels in the western U.S. and higher levels in the eastern U.S. The eastern U.S. is estimated to have more natural organic fine-mode particles and more water associated with hygroscopic fine-mode particles than the western U.S. due to generally higher humidity levels.

Analysis of PM Measurements from the Improve Monitoring Network

The Interagency Monitoring of Protected Visual Environments (IMPROVE) program is a cooperative visibility monitoring effort among the EPA, federal land management agencies, and state air agencies. One of the functions of this program is to monitor visibility and aerosol conditions in Class I areas, and for the most part the IMPROVE monitors are located in rural areas.

An estimate of the range of background concentrations on a daily basis can be obtained from reviewing multi-year data at remote locations. EPA staff have conducted an analysis of daily $PM_{2.5}$ measurements from 1990 to 2002 at IMPROVE sites across the U.S., focused on the non-sulfate components of $PM_{2.5}$. Data collected before 1990 were not used due to a sulfate analytical problem before March 1989[2]. Ambient sulfate concentrations are almost entirely due to anthropogenic sources, so while non-sulfate $PM_{2.5}$ is partly of anthropogenic origin, it captures almost all of the background. For this analysis we calculate daily non-sulfate $PM_{2.5}$ concentrations according to equation 1 (Malm et al., 2000).

$$\text{non-sulfate } PM_{2.5} = PM_{2.5} - 4.125*S, \qquad (1)$$

where $PM_{2.5}$ is the measured mass of fine PM and S is the measured mass of fine sulfur. This equation assumes that all elemental sulfur measured is from sulfate, and that all sulfate is ammonium sulfate. If some of the sulfate were ammonium bisulfate, then this would overestimate sulfate mass, since ammonium bisulfate is lighter than ammonium sulfate. For example, if as much as 16 percent of the sulphate were ammonium bisulfate and 84 percent were

2 http://vista.cira.colostate.edu/improve/Data/QA%5FQC/Issues/SO4underreported.htm

ammonium sulfate, then the correct multiplier in equation 1 would be 101/25, and we would be overestimating sulfate by two percent.

The sulfur concentration measurements are sometimes biased low (by as much as a factor of two) at Eastern sites under high humidity conditions. We do not try to adjust for this bias. For a discussion of this problem see *Underestimation of Sulfur Concentrations During High Loadings and Humidity Conditions in the Eastern U.S.* at http://vista.cira.colostate.edu/improve/Data/QA_QC/Issues/S_underestimation.htm.

Data Completeness Requirements

Before calculation of statistics, we impose data completeness requirements to reduce bias due to imbalances in the numbers of seasonal measurements. We require that each calendar quarter for each site must have least 11 observations; otherwise that quarter is dropped. We then further subset the data to years with all seasons for each monitor, and only use monitors with at least three years of data.

Long-Term Means

Figure 1 presents the long-term means of non-sulfate $PM_{2.5}$ measurements at all of the IMPROVE sites satisfying the above data completeness requirements. From inspection of this figure we see that most long-term means in the northeast are below 4 $\mu g/m^3$. In the southeast the means range from 4 to almost 10 $\mu g/m^3$. A major contribution to $PM_{2.5}$ concentrations in this region is from anthropogenic emissions, and we cannot at this time separate those out from background. Except for California, Oregon, and Washington, sites in the western half of the U.S. all have means below 4 $\mu g/m^3$ save for one site in the 4-5 $\mu g/m^3$ range. Some of the higher concentrations observed in these states are partially due to wildfires, prescribed fires, wood burning, and dust storms.

Daily Variability

Figure 2 illustrates the variability (standard deviations) of the daily non-sulfate $PM_{2.5}$ concentrations at the IMPROVE sites. We recognize that contributing to this variability are (1) fluctuations of anthropogenic concentrations, (2) highly variable exceptional natural events, and (3) the fluctuations in background. We will not attempt to separate these components at this time. The variability of annual means is not included: the standard deviations plotted are the within-year standard deviations (the means of the standard deviations for each year, for each monitor).

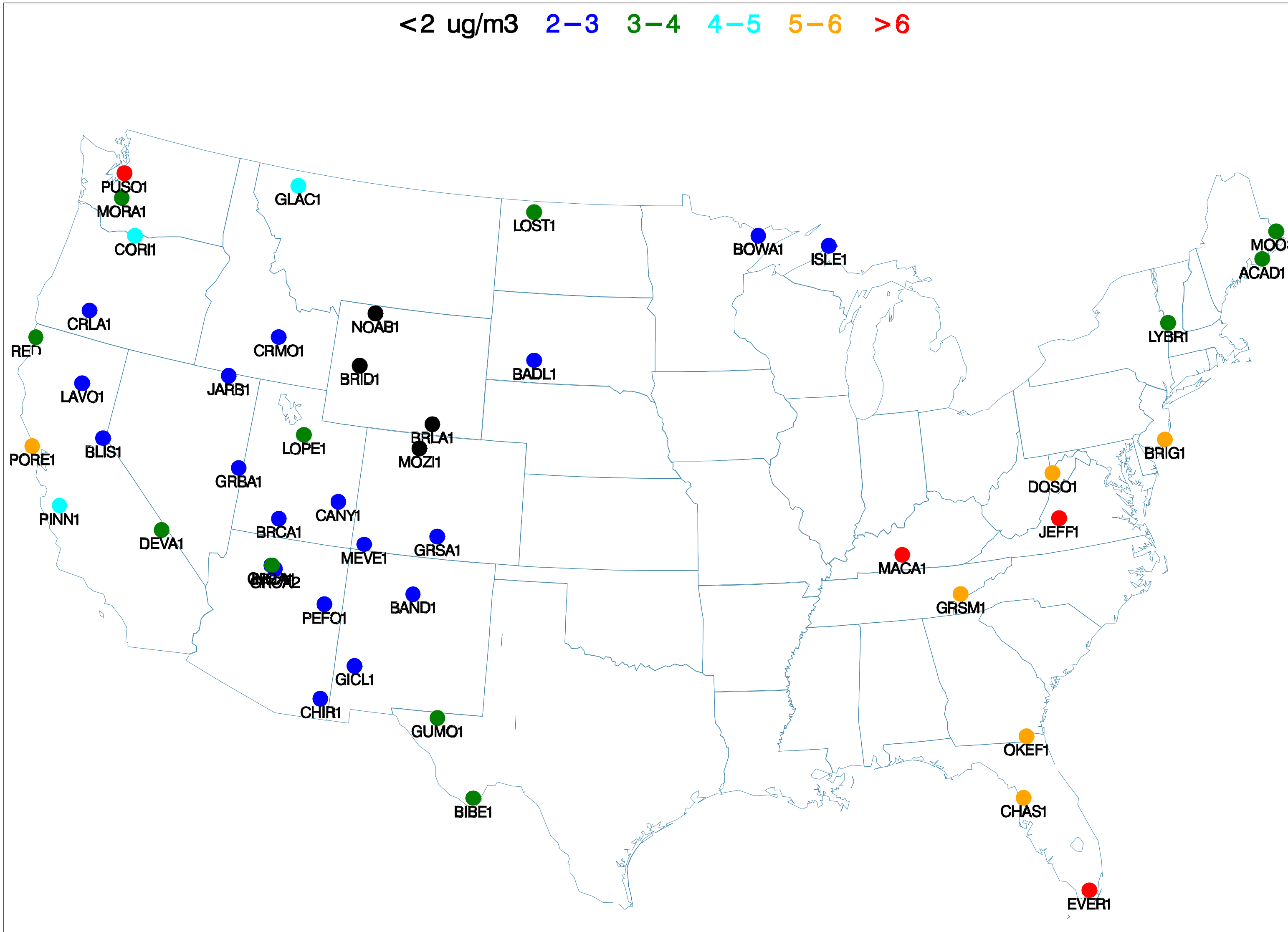

Figure 1. Long-term means of non-sulfate $PM_{2.5}$ at IMPROVE sites

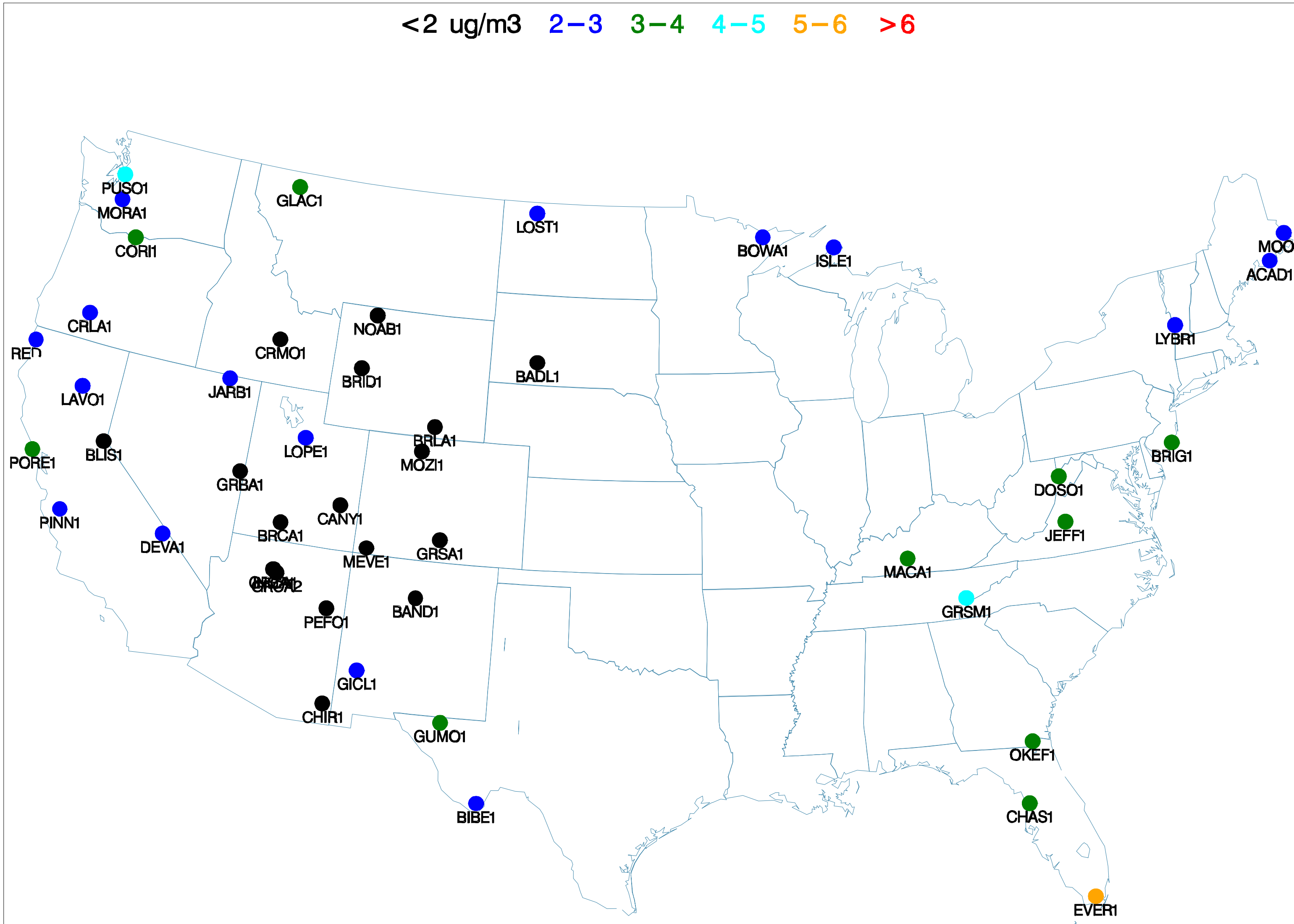

Figure 2. Long-term standard deviations of non-sulfate $PM_{2.5}$ at IMPROVE sites

There is only one site in the U.S. with standard deviation of daily non-sulfate $PM_{2.5}$ concentrations above 5.2 μg/m^3, Sequoia National Park in California (SEQU), with a standard deviation of 6.8 μg/m^3. This site is heavily impacted by fires and also is affected by transport of pollutants from the San Joaquin Valley. The standard deviations in the large area of the West not including California and Washington are all below 3 μg/m^3, with two exceptions, Glacier National Park, Montana (GLAC) and Guadalupe Mountains National Park, Texas (GUMO), which have standard deviations of 3.7 and 3.2 μg/m^3 respectively.

The long-term means and variability at sites in these broad regions are summarized in Table 2. We are using the term "daily standard deviations" to refer to the standard deviations of the daily concentrations. The regions specified in Table 2 are illustrated in Figure 3.

Table 2. Regional averages and ranges of long-term means and daily standard deviations of non-sulfate $PM_{2.5}$ concentrations at IMPROVE sites (μg/m^3)

Region	States in region	Means	St Devs	# sites
Alaska	Alaska	1.2	1.5	1
Central West	AZ, CO, ID, KS, MT, ND, NE, NM, NV, OK, SD, TX, UT, WY	2.5 (1.6-4.6)	1.9 (1.3-3.7)	37
East-Southeast	AL, AR, DC, DE, FL, GA, IA, IL, IN, KY, LA, MD, MO, MS, NC, NJ, OH, PA, SC, TN, VA, WV	6.1 (4.2-9.5)	3.7 (2.9-5.2)	14
Great Lakes	MI, MN, WI	2.7 (2.5-3.0)	2.4 (2.2-2.7)	4
Hawaii	Hawaii	1.1 (0.7-1.8)	0.9 (0.8-1.0)	3
New England	CT, MA, ME, NH, NY, RI, VT	3.3 (3.1-3.6)	2.5 (2.1-2.8)	3
West coast	CA, OR, WA	4.3 (2.2-8.6)	3.2 (1.8-6.8)	16

Note: The "Means" column has the mean of the long-term averages of the sites in the region followed by the minimum and maximum of the long-term averages of the sites in the region in parentheses. Similarly for the "St Devs" column, which presents standard deviations of the daily concentrations about the annual means.

The larger means and standard deviations in the southeast (compared to the northeast) may be partly due to a higher frequency of forest fires and agricultural dust.

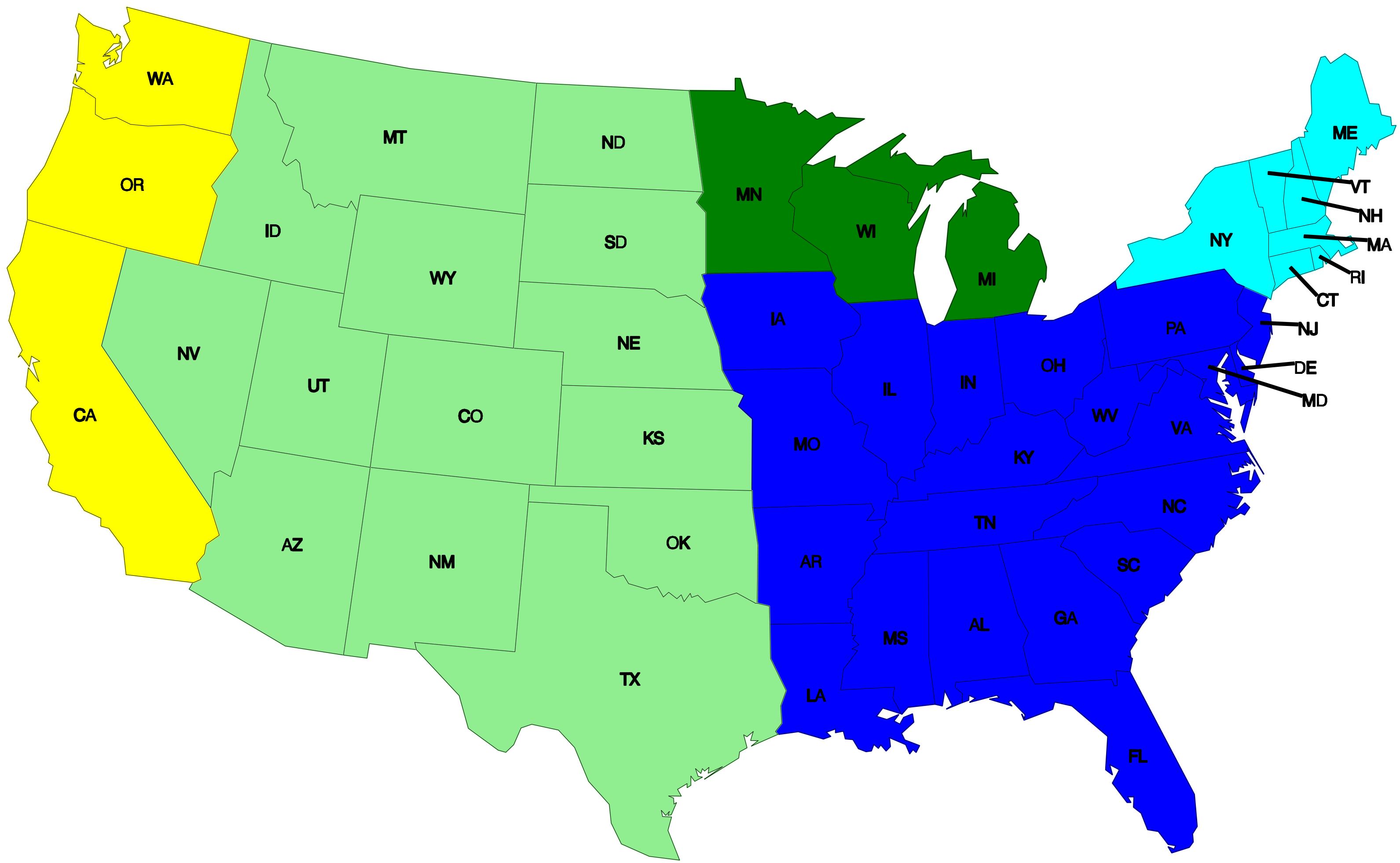

Figure 3. Broad Regions (Alaska and Hawaii not shown)

Estimates of Background Concentrations of $PM_{2.5}$

The regions in Figure 3 were selected based on geography and similar concentrations of non-sulfate $PM_{2.5}$. Clearly the sites in the southeast region all see significant impacts of anthropogenic pollution, and levels of anthropogenic pollution are not relevant to a selection of regions with similar background concentrations. Therefore, based on regional differences in geography and land use, we divide the U.S. into a number of regions for estimating regional background levels (Table 3). The "Eastern U.S." region includes Minnesota, Iowa, Missouri, Arkansas, Louisiana, and states east of these states. The "Central West" region comprises states west of the Eastern U.S. region and east of Washington, Oregon, and California. Washington, Oregon, and northern California make up the "North West Coast" and California south of about 40 degrees latitude the "South West Coast" regions. Alaska and Hawaii each are taken to be a separate region. These regions are shown in Figure 4.

Table 3. Background regions

Region	Characteristics
Eastern U.S.	More vegetation, higher rainfall
Central West	Less vegetation and rainfall
North West Coast	Pacific coast, higher rainfall
South West Coast	Pacific coast, less rainfall
Alaska	Northern
Hawaii	Islands in Pacific

To determine estimates of background we use the averaged measured non-sulfate $PM_{2.5}$ values at IMPROVE sites in these regions. The Eastern U.S. region is heavily impacted by anthropogenic emissions and we selected sites in northern states, which we judge to be affected to a lesser extent by anthropogenic pollution, to form estimates of background concentrations, using all IMPROVE sites in the selected states. In all of the other regions we include all of the IMPROVE sites. Table 4 describes the IMPROVE sites selected to represent these different regions of the U.S. We recognize that these estimates will be too high, as they include an anthropogenic component, some sites more than others.

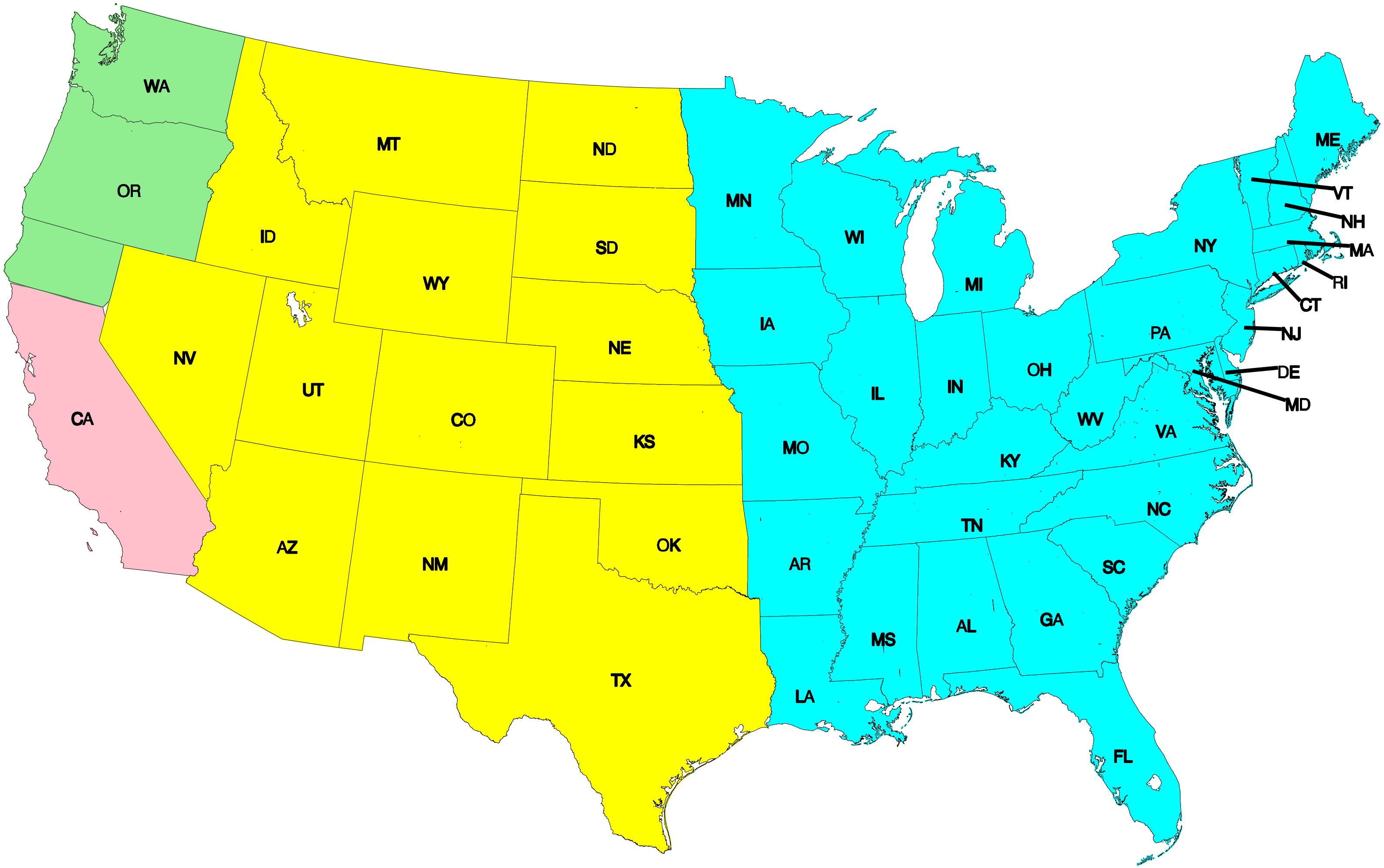

Figure 4. Regions for Background Estimates (Alaska and Hawaii not shown)

Table 4. IMPROVE sites selected for estimates of regional background

Region	IMPROVE sites
Eastern U.S.	All sites in Maine, New Hampshire, Vermont, Minnesota, and Michigan
Central West	All sites in this region (Idaho, Montana, Wyoming, North Dakota, South Dakota, Colorado, Utah, Nevada, and Arizona)
North West Coast	All sites in this region (all Washington and Oregon sites, and the northern California sites REDW and LAVO)
South West Coast	All sites in this region (all California sites except the northern sites REDW and LAVO)
Alaska	All sites in Alaska
Hawaii	All sites in Hawaii

The 99th percentile concentrations at each of these sites were calculated to assess high values measured at these sites, while avoiding excursions that potentially reflect exceptional natural events. Standard deviations were also calculated for characterization of the daily variation of background concentrations. Table 5 presents the results of this analysis as means and ranges of individual site statistics within each of the background regions. Although these represent upper-bound estimates of background concentrations of $PM_{2.5}$, the distributions of daily non-sulfate $PM_{2.5}$ concentrations at these sites provide an indication of the ranges for the daily variability of $PM_{2.5}$ background concentrations, and the 99th percentiles of these distributions are an estimate of the highest daily background concentrations.

Table 5. Estimates of long-term means, daily standard deviations, and 99th percentiles of $PM_{2.5}$ background concentrations ($\mu g/m^3$)

Region	# sites	Means	St Devs	99^{th} %iles
Eastern U.S.	7	3.0 (2.5-3.6)	2.5 (2.1-2.8)	13 (11-15)
Central West	37	2.5 (1.6-4.6)	1.9 (1.3-3.7)	10 (6-17)
North West Coast	8	3.4 (2.2-6.6)	2.8 (2.1-4.2)	14 (10-21)
South West Coast	8	5.2 (2.6-8.6)	3.7 (1.8-6.8)	20 (9-33)
Alaska	1	1.2	1.5	9
Hawaii	3	1.1 (0.7-1.8)	0.9 (0.8-1.0)	4 (4-5)

Notes:
1) Some of these estimates likely contain a significant North American anthropogenic component.
2) The "Means" column has the mean of the long-term averages of the sites representing the region followed by the minimum and maximum of the long-term averages of these sites in parentheses. Similarly for the "St Devs" column, which presents standard deviations of the daily concentrations about the annual means, and the "99^{th} %iles" column, which presents the 99^{th} percentiles of the daily concentrations over the 23-year period.

Impact of High-PM Episodes

We make a distinction between baseline and episodic background levels, and the values in Table 2 include both of these components. We have performed analyses of specific short-term episodes of high non-sulfate $PM_{2.5}$ concentrations where we identify the time periods that sites are clearly impacted and calculate means and standard deviations with and without those time periods removed from the data. Table 6 summarizes the results of these analyses and shows the impact of each episode on the annual means and standard deviations of non-sulfate $PM_{2.5}$ concentrations for the affected sites. Time series graphs of the non-sulfate $PM_{2.5}$ measurements for selected episodes and sites in Table 6 are presented in Figures 5 through 9.

Table 6. Impacts of episodes of high non-sulfate $PM_{2.5}$ concentrations on annual means and daily variability ($\mu g/m^3$)

Event	Site	Episode dates	Effect of dropping episode on annual mean	Effect on standard deviation	Maximum 24-hour concentration in episode
2002 Quebec fires	BRIG, NJ	7/7/02	6.1 to 5.2 Δ=0.9 (15%)	9.8 to 3.5 Δ=6.3 (64%)	101
2002 Quebec fires	LYBR, VT	7/7/02	3.7 to 2.8 Δ=0.9 (23%)	7.1 to 3.0 Δ=4.1 (58%)	68
2002 Quebec fires	QURE, MA	7/7/02	3.9 to 3.4 Δ=0.5 (13%)	6.2 to 2.7 Δ=3.5 (56%)	65
Single day high concentration (unknown origin)	GUMO, TX	3/24/02	4.0 to 3.5 Δ=0.5 (11%)	5.5 to 2.6 Δ=2.9 (53%)	56
Single day high concentration (unknown origin)	CHIR, AZ	10/16/01	2.48 to 2.24 Δ=0.24 (10%)	3.15 to 1.85 Δ=1.3 (41%)	30
Single day high concentration (unknown origin)	CHIR, AZ	4/8/00	3.0 to 2.8 Δ=0.2 (7%)	3.1 to 2.0 Δ=1.1 (36%)	27
May 1998 Central American fires (Tanner et al., 2001)	GRSM, TN	May 11-30, 1998	6.24 to 5.8 Δ=0.44 (7%)	4.47 to 3.64 Δ=0.83 (18%)	27
May 1998 Central American fires	SIPS, AL	May 11-30, 1998	8.24 to 7.66 Δ=0.58 (7%)	4.37 to 3.77 Δ=0.6 (14%)	19

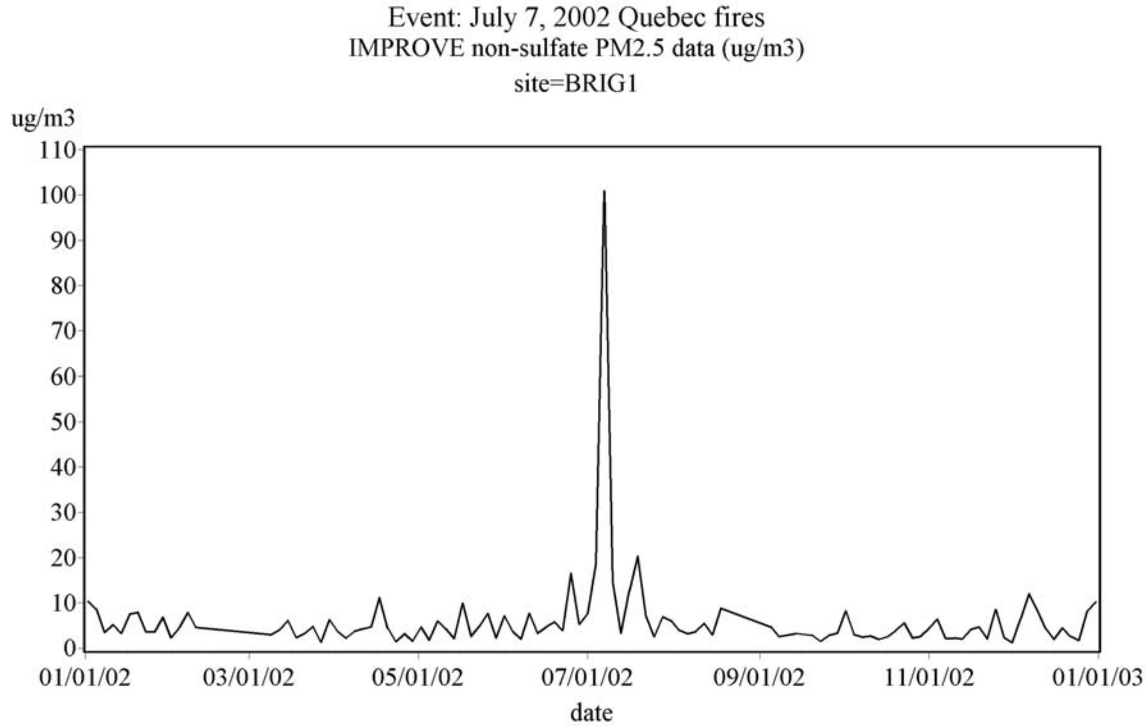

Figure 5. 2002 Quebec fires, Brigantine National Wildlife Refuge, NJ site

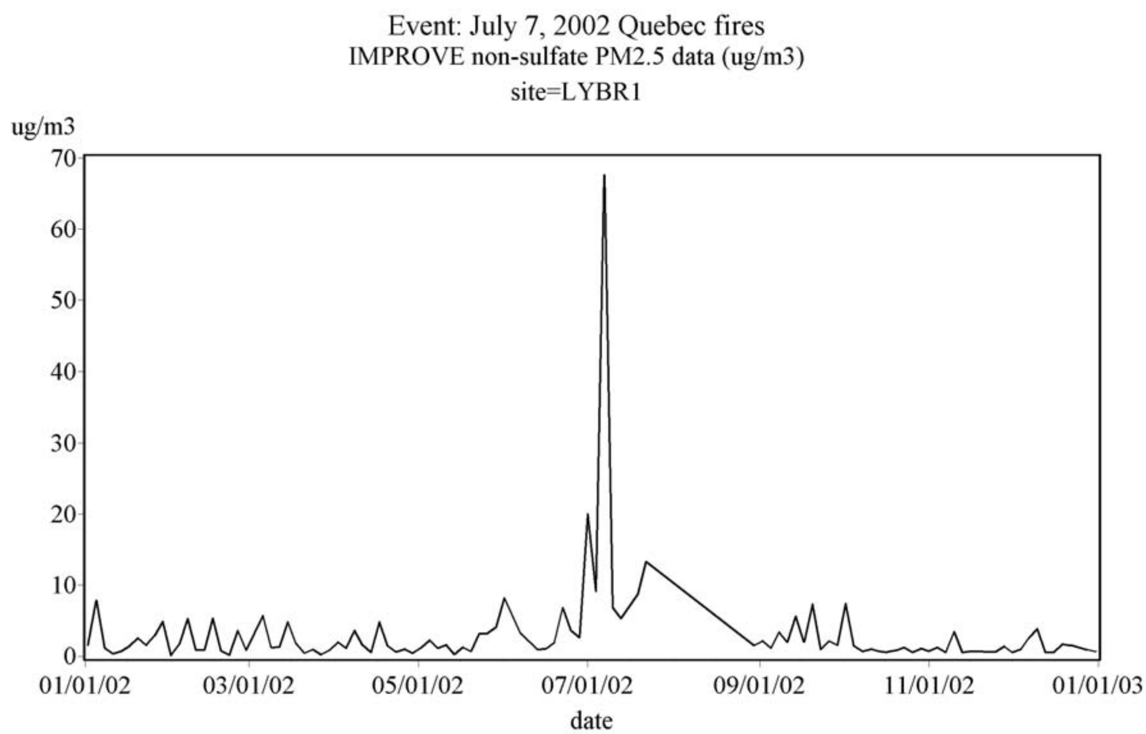

Figure 6. 2002 Quebec fires, Lye Brook Wilderness, VT site

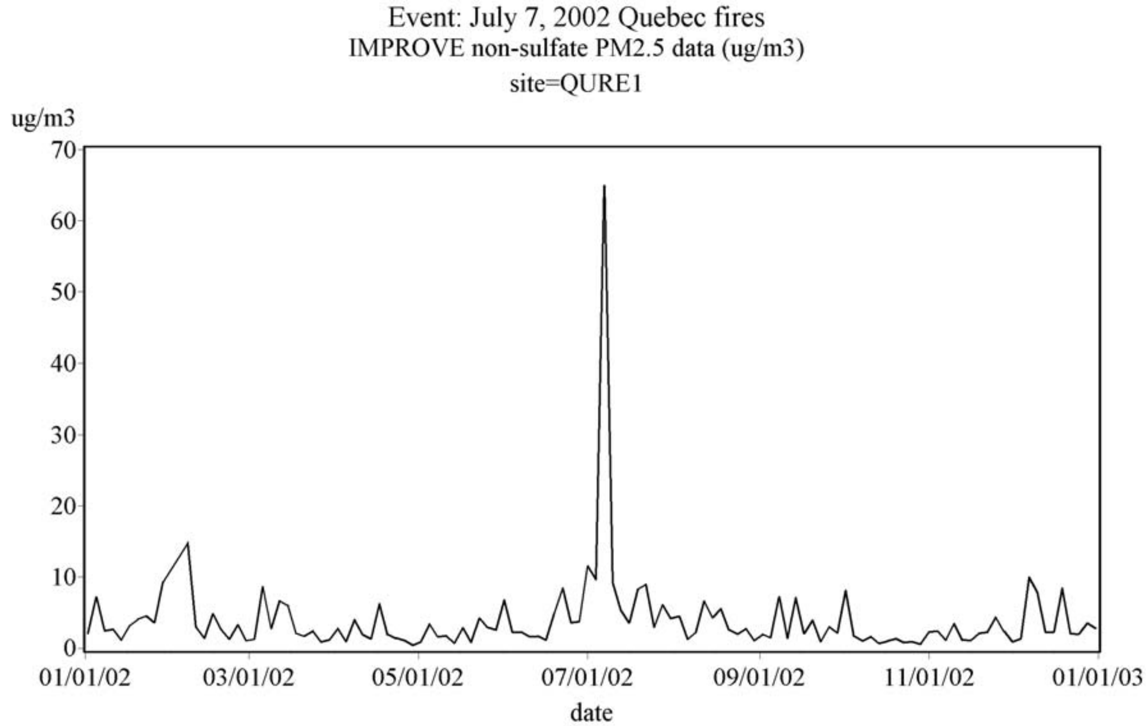

Figure 7. 2002 Quebec files, Quabbin Summit, MA site

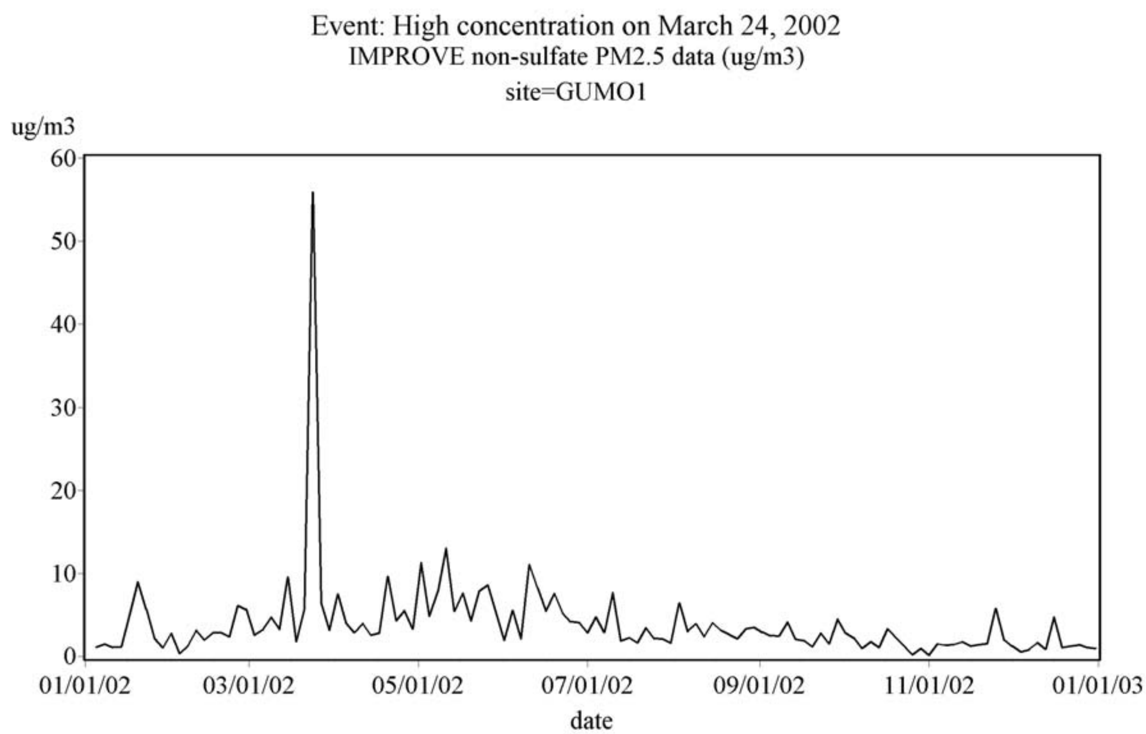

Figure 8. 2002 high concentration event, Guadalupe Mountains National Park, TX site

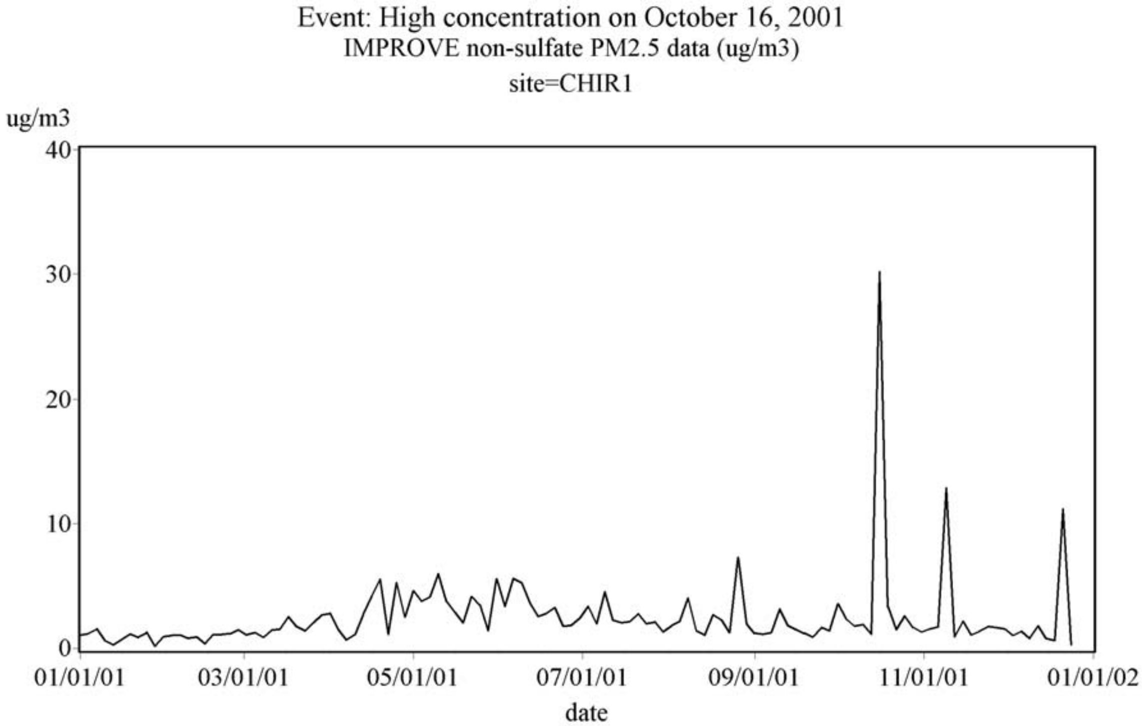

Figure 9. 2001 high concentration event, Chiricahua National Monument, AZ site

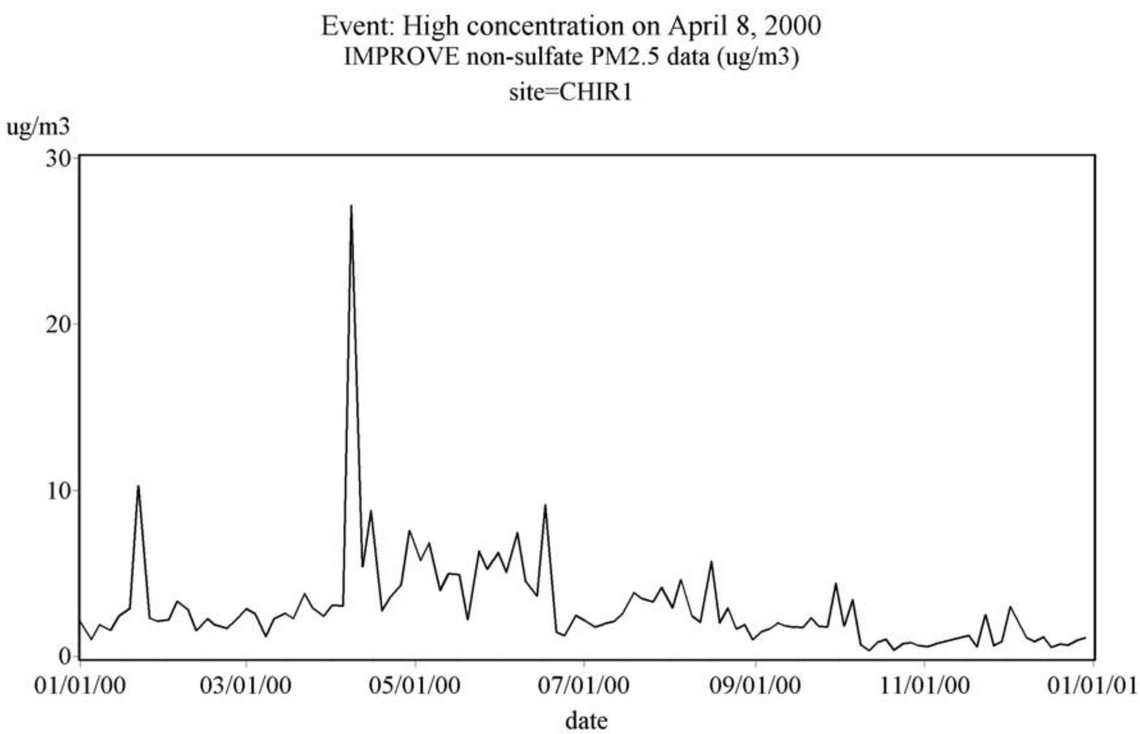

Figure 10. 2000 high concentration event, Chiricahua National Monument, AZ site

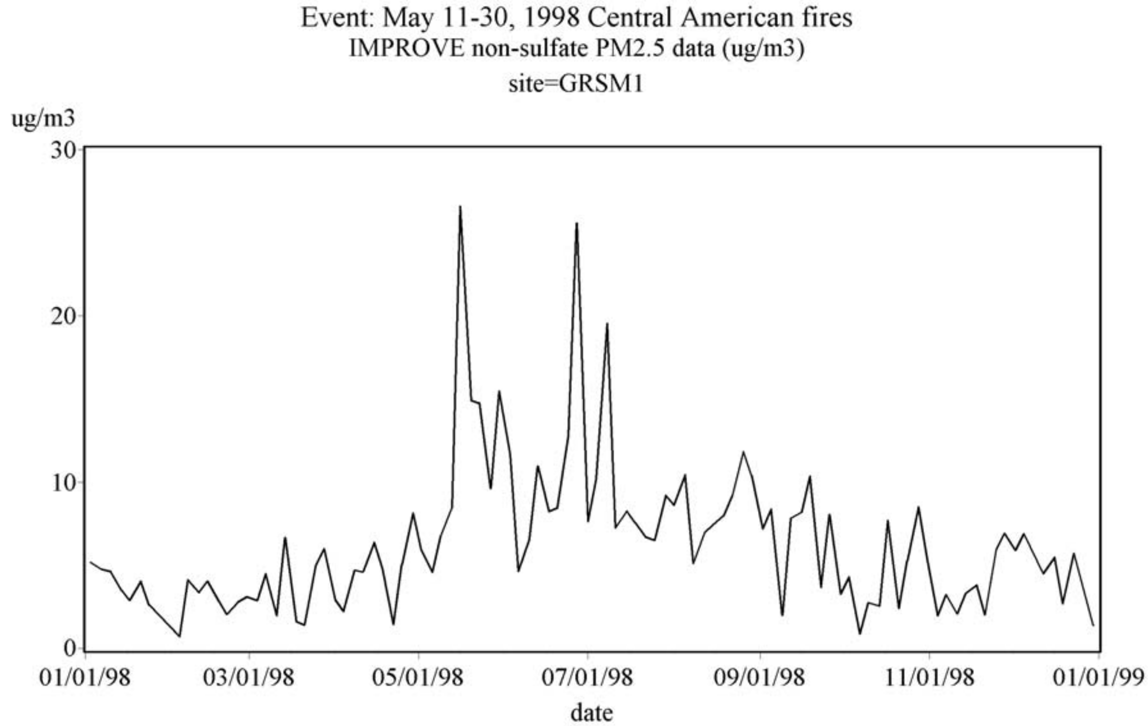

Figure 11. 1998 Central American fires, Great Smoky Mountains National Park, TN site

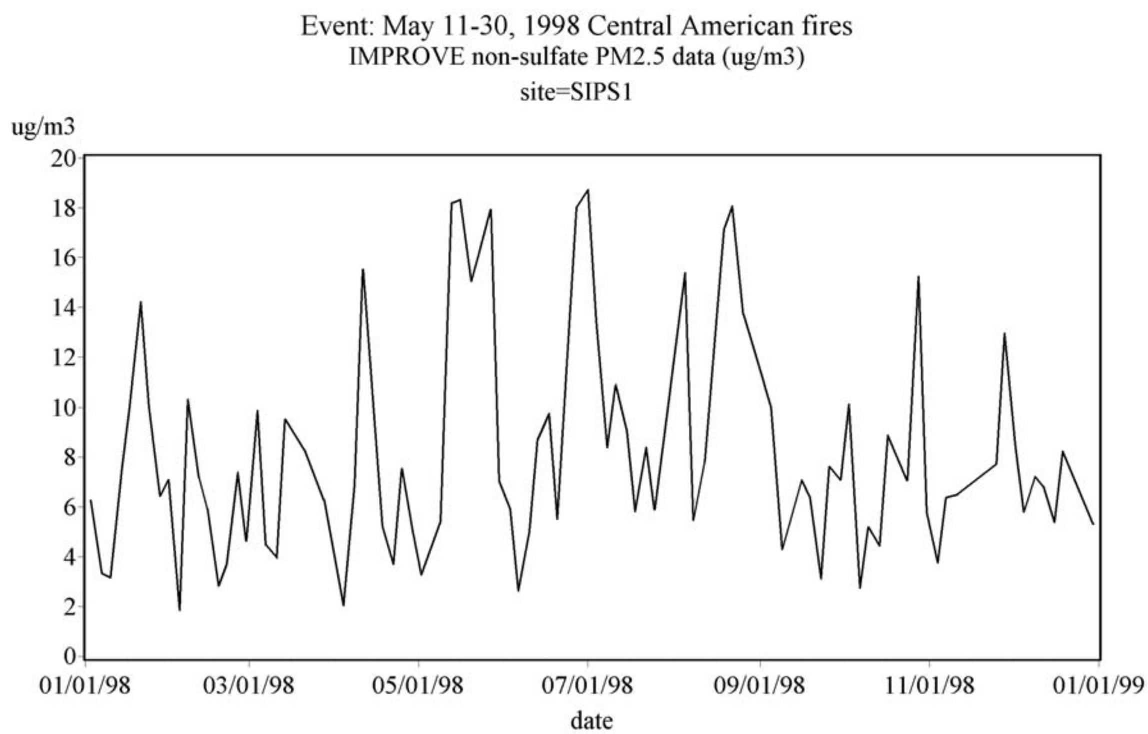

Figure 12. 1998 Central American fires, Sipsy Wilderness, AL site

References

EPA (1996). Air Quality Criteria for Particulate Matter. EPA Office of Research and Development, Research Triangle Park, NC 27711. EPA/600/P-95/001aF-cF.

EPA (2004). Air Quality Criteria for Particulate Matter. EPA National Center for Environmental Assessment, Research Triangle Park, NC 27711. EPA/600/P-99/002aD.

EPA (2005). Review of the National Ambient Air Quality Standards for Particulate Matter: Policy Assessment of Scientific and Technical Information, Second Draft PM Staff Paper. EPA Office of Air Quality Planning and Standards, Research Triangle Park, NC 27711.

Malm, W. C., et al. (2000) Spatial and seasonal patterns and temporal variability of haze and its constituents in the United States. Report III. Fort Collins, CO: Cooperative Institute for Research in the Atmosphere, Colorado State University. Available: http://vista.cira.colostate.edu/improve/Publications/Reports/2000/2000.htm

Tanner, R., et al. (2001) Impact of the 1998 Central American fires on $PM_{2.5}$ mass and composition in the southeastern United States. Atmospheric Environment 35 (2001) 6539-6547.